Mesoscale Dispersion of ^{85}Kr in the Vicinity of the AREVA La Hague Reprocessing Plant

O. Connan[1,*], K. Smith[2], L. Solier[1], D. Maro[1], D. Hébert[1], and G. Bacon[1]

[1] *IRSN/DEI/SECRE, Laboratoire de Radioécologie, Cherbourg-Octeville, France*
[2] *RPII, Radiological Protection Institute of Ireland, Clonskeagh, Dublin 14, Ireland*
* *Corresponding author: olivier.connan@irsn.fr*

Abstract. The Institut de Radioprotection et de Sureté Nucléaire (IRSN) performed a series of air sampling campaigns at mesoscale distances (10 - 80 km) from the AREVA La Hague reprocessing plant (north west of France) between 2007 and 2009. These samples were collected in order to test and optimise a technique to measure low krypton-85 (^{85}Kr) air concentrations, and to investigate the performance of three atmospheric dispersion models (RIMPUFF, HySplit, and ADMS). This paper presents ^{85}Kr air concentrations measured at both land and sea locations. In addition, this paper compares the measured ^{85}Kr air concentrations, which varied from 2 to 8000 Bq m^{-3}, with the predictions from the atmospheric dispersion models. During stable wind conditions, the dispersion models make reasonable estimates of the ^{85}Kr field measurements. In contrast, the models fail to accurately predict temporal peaks in concentration during periods of rapid and large changes in wind speed and/or wind direction.

1. INTRODUCTION

Atmospheric dispersion models (ATDMs) predict the movement and deposition of radionuclides in the environment. This can be a useful tool in the early phases of an emergency, following an unplanned atmospheric release of radionuclides when field measurements may not yet be available. Dispersion models can be used to estimate the radiation doses received by the public and also predict the geographic areas affected. This information is an essential aid to decision makers when deciding on appropriate countermeasures for the protection of the public and on the deployment of monitoring teams in the field [1,2]. Thus, it is vital to have an understanding of the limitations and uncertainties of the model so that results can be interpreted and reported with confidence. One approach to assess the performance of ATDMs involves validation, i.e. comparing predictions from the ATDMs with measurements made in the field [3].

Krypton-85 (^{85}Kr), a radioactive noble gas with a half life of 10.7 y, is released into the atmosphere primarily during the reprocessing of spent nuclear fuel. The AREVA La Hague reprocessing plant (North Cotentin peninsula, France) discharges ^{85}Kr from two production units (UP2-800 and UP3) [4]. In the vicinity of this source, high ^{85}Kr air concentrations allow real-time measurements to be made. At distances far removed from the source or where there is a relatively low ^{85}Kr air concentration, a pre-concentration step may be

required in the sample analysis. The ^{85}Kr discharges from this plant provide a well defined source term for use in validating ATDMs.

In this study, the IRSN performed a series of air sampling campaigns at mesoscale distances (10 - 80 km) from the reprocessing plant. These samples were collected in order to: (i) test and optimise the ^{85}Kr measurement technique at low concentrations, (ii) investigate the performance of ATDMs by comparing the ^{85}Kr air concentrations predicted by the models with IRSN measurements and, (iii) assess the ability of ^{85}Kr to be used as a tracer of dispersion processes at mesoscale distances. The results of the ^{85}Kr concentration measurements and a comparison with the predictions of the ATDMs are presented and discussed in this paper.

2. METHODOLOGY

2.1 Krypton-85 measurement techniques

2.1.1 Real-time ^{85}Kr measurements

At close range (< 20 km) to the discharge point, the ^{85}Kr air concentration is usually sufficiently high to allow real-time measurements. In these circumstances, the ^{85}Kr activity may be determined by beta counting an air sample in a gas proportional counter. The IRSN operate a Berthold-LB111 proportional counter. In this detector a pump circulates air in a measurement chamber that is surrounded by four LB123 proportional counters. The LB111 has a 500 Bq m^{-3} detection limit for a 1 minute counting time [5].

2.1.2 Low ^{85}Kr air concentration measurements

A pre-concentration step in the sample analysis is required when the ^{85}Kr air concentration is low (between 1 and 500 Bq m^{-3}). This is usually the case at distances greater than 30 km from the source. The pre-concentration involves first collecting a 4 m^3 sample of air. The ^{85}Kr in the air (as well as some nitrogen, oxygen, and other trace atmospheric elements) is then trapped onto activated charcoal maintained at the temperature of liquid nitrogen and at reduced pressure. The ^{85}Kr is then released by heating the charcoal column to 300°C and collected in a 15 l sample bag. This 15 l is subsequently compressed to 5 bar into a suitable counting geometry. The sample activity is then quantified by counting the 514 keV gamma emitted during ^{85}Kr decay using an ORTEC HPGe detector. Stable Kr, obtained by Gas Chromatography/Mass Spectrometry (GC/MS) measurement (Agilent 5973 GC/MS) before and after the Kr concentration steps, is used as a yield tracer for the concentration process. The limit of detection with this method of enrichment combined with gamma spectrometry (1 to 5 day count period) is approximately 1.5 Bq m^{-3}.

2.2 Atmospheric dispersion models

2.2.1 HySplit

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HySplit) model simulates the dispersion of a pollutant by assuming either a puff or particle dispersion. Advection and diffusion calculations are made in a Lagrangian framework while concentrations are calculated on a fixed (Eulerian) grid [6]. HySplit requires gridded meteorological data defined at regular time intervals. The meteorological data used in this work was created by the Global Data Assimilation System (GDAS) and has one degree horizontal grid spacing. All simulations were performed using a 3D particle simulation with two discharge points (UP2-800 and UP3) and 100 m release heights. The model used a nominal

release rate of 1 Bq hr^{-1}. The ^{85}Kr air concentration values calculated by the model and output every 10 min were scaled according to the average ^{85}Kr discharge rate (Bq hr^{-1}) during the period modelled.

2.2.2 ADMS

ADMS is an advanced Gaussian dispersion model developed by Cambridge Environmental Research Consultants [7] and recommended by US-Environmental Protection Agency. This model is used to predict the atmospheric dispersion and concentration of pollutants released by industrial sources. Model calculations assume stable and neutral weather conditions and dispersion is as a function of the characteristics of the atmospheric boundary layer (e.g. Monin-Obukhov length, boundary layer height). The model also considers the site characteristics (buildings, roughness, topography). In this work, ADMS was setup in simple configuration with only ^{85}Kr discharge data and meteorology. Results were output every hour for a fixed point corresponding to the (IRSN) measurement points.

2.2.3 RIMPUFF

The RIsø-Mesoscale-PUFF (RIMPUFF) model is a Lagrangian mesoscale atmospheric dispersion model. RIMPUFF simulates the release of radioactivity using a series of consecutively released puffs. Each puff is released with a fixed amount of radioactive material over a specified grid. At each time step the model advects, diffuses and deposits the individual puffs according to local meteorological parameter values [8]. RIMPUFF is driven by meteorological data created by the High Resolution Limited Area Model (HIRLAM). In this work, RIMPUFF used HIRLAM data with a 40 km horizontal resolution provided by the Irish Meteorological Service (Met Éireann). All simulations assumed a single (100 m high) source at the La Hague site. RIMPUFF was initiated with hourly ^{85}Kr discharge data provided by AREVA for the sampling dates. Model output consisted of instantaneous ^{85}Kr air concentrations (Bq m^{-3}) calculated every 10 mins at the (IRSN) measurement points.

3. EXPERIMENTAL AREA

The AREVA La Hague plant is located on the North Cotentin peninsula. This is a narrow (5 km wide) and hilly area that is bounded by steep cliffs on the south of the site. The vegetation consists mainly of moors and hedged farmland. The plant rises to a height of 180 m above sea level, and contains two production units (UP2-800 and UP3) each with a 100 m high stack that are 200 m apart. When nuclear fuel reprocessing is in progress, ^{85}Kr is discharged into the environment from the plant stacks at intervals of between 30 and 45 min.

In the course of this work, the IRSN performed three ^{85}Kr measurement campaigns, two on land (Doville and Le Laboratoire de Radioécologie de Cherbourg-Octeville (LRC)) and one at sea (DISPAMER campaign). All the sampling sites (Figures 1 and 4) are situated around the North Cotentin peninsula, at distances between 18 and 80 km from the AREVA La Hague plant.
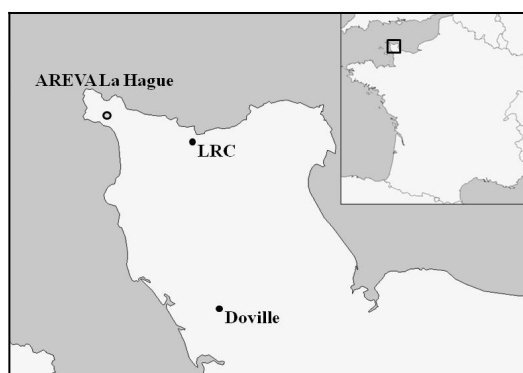
Figure 1. Locations of the land based sampling sites (Doville and LRC) and the AREVA La Hague plant.

4. RESULTS AND DISCUSSION

4.1 Doville

On 30th January 2008, four air samples were retrieved at Doville for ^{85}Kr analysis. Doville is located 47 km south-southeast of the AREVA La Hague plant (the facility is located at 330° relative to Doville). Wind speed and direction (Figure 2 (B)) for the AREVA La Hague site show that during the sampling period the wind speeds ranged from 7 to 9 m.s^{-1}, regularly changing directions: backing from 350° to 310° during the sampling period.

The IRSN Kr-85 measurements are shown in Figure 2 (A) along with the model predictions of air concentration. There is good correlation between the measured ^{85}Kr values and those predicted by HySplit and RIMPUFF. The ADMS model poorly predicts the temporal location of the peak value. HySplit overestimates the peak concentration (273 Bq m^{-3}) and ADMS underestimates the peak concentration (peak value 10 Bq m^{-3}). RIMPUFF gives the best correlation with the measured results, predicting a peak concentration of 131 Bq m^{-3} compared to the measured concentration of 136 Bq m^{-3}.
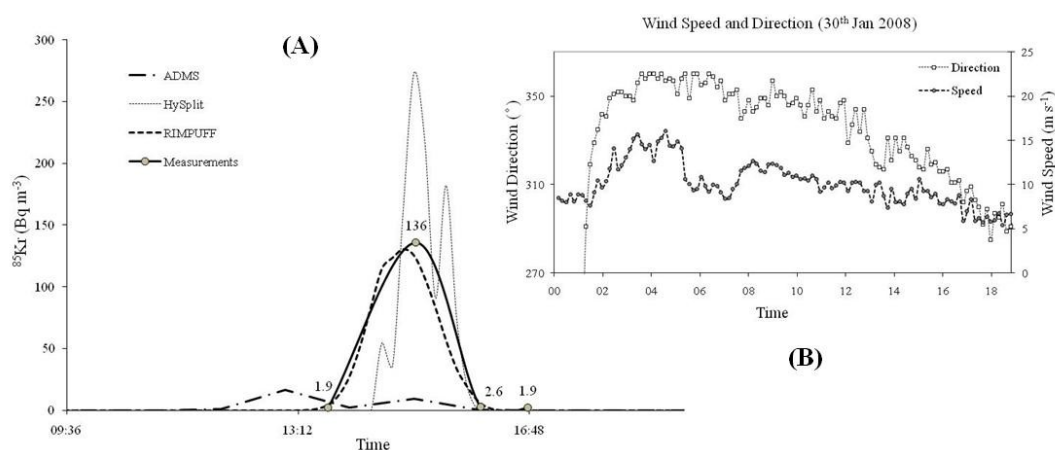

Figure 2. The ^{85}Kr measurements (Bq m^{-3}) recorded at Doville on 30th January 2008 compared with results of model simulations (A) and wind speed and direction recorded at the reprocessing plant (shown in B).

4.2 LRC

On the 2nd February 2009, real-time measurements of ^{85}Kr were performed at the LRC site. The 10 min averaged ^{85}Kr concentrations are presented together with model predictions in Figure 3 (A). This time series shows a measured ^{85}Kr peak concentration of 7000 Bq m^{-3} which occurred between 20:20 and 20:45. A second, smaller peak concentration of 2000 Bq m^{-3} occurred between 21:45 and 21:55. Both HySplit and RIMPUFF were close in predicting the temporal location of the first peak ^{85}Kr concentration compared to the ADMS model. All three models failed to predict the second peak concentration. Both HySplit and ADMS grossly underestimated the peak activity concentration and the peak activity concentration predicted by RIMPUFF is approximately one third that of the measured value.

Wind speed and direction (Figure 3 B) for the AREVA La Hague site show that the wind blew in the direction of the LRC between 19:30 and 20:40 and then again between 21:10 and 21:40 (LRC site is located at 286° from the AREVA plant). Allowing for plume travel, these time periods correspond to the two IRSN measured peak concentrations at 20:40 and 21:50. The temporal resolution of the HIRLAM and GDAS met data are not sufficient to model the wind direction change between 21:00 and 22:00. This would explain why the smaller second peak is not predicted by the models.
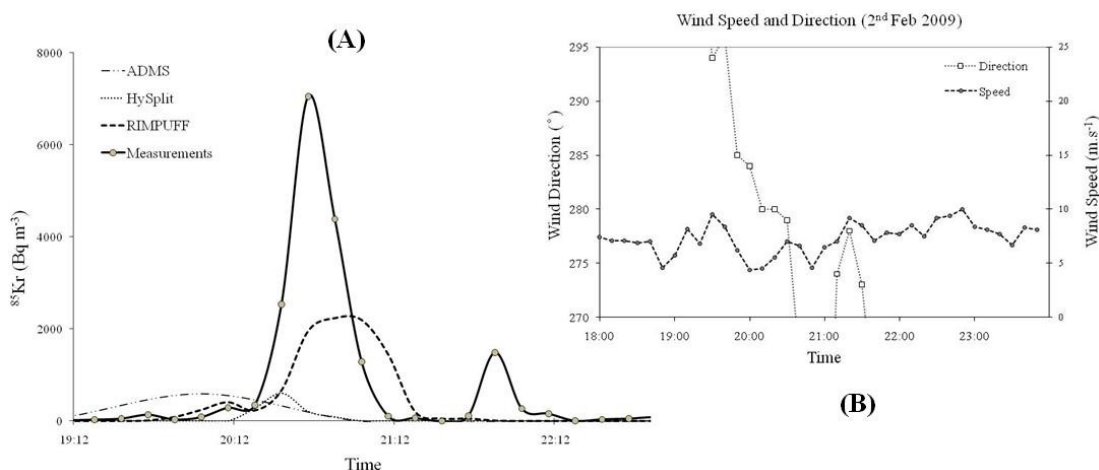

Figure 3. The ^{85}Kr measurements (Bq m^{-3}) recorded at LRC on 2nd February 2009 compared with results of model simulations (A) and wind speed and direction recorded at the reprocessing plant (shown in B).

4.3 DISPAMER Campaign

Winds travelling over the AREVA La Hague site are rarely directed over land thus making it difficult to establish land based sampling. The IRSN carried out a sea campaign (DISPAMER) in the Western English Channel (27th September to 2nd October 2008) in an attempt to retrieve a number of samples with above-background ^{85}Kr concentrations. During the DISPAMER campaign, the RIMPUFF model was run twice daily to predict the direction of the plume over the following 48h. These forecasts were used to position the ship in the plume's predicted path for air sampling.

Unfortunately, due to a combination of poor weather conditions which prevented the ship from getting to the required sampling locations and low discharges (~ 4 TBq hr^{-1}) from the reprocessing plant during the sampling

campaign, only a small number of samples with [85]Kr concentrations above background were collected. Sampling locations and significant results are shown in Figure 4 along with the RIMPUFF and HySplit model predictions.
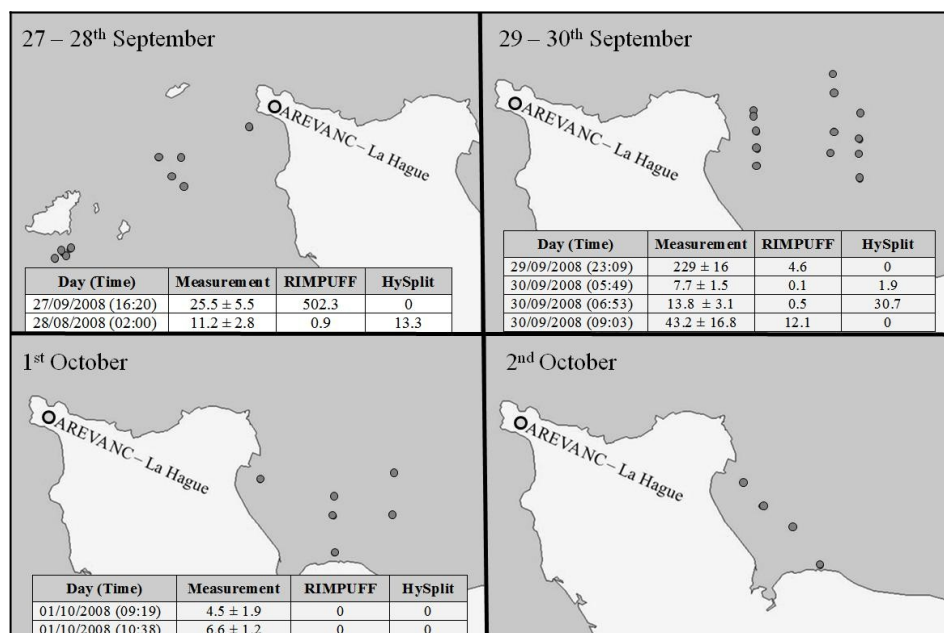

27 – 28th September

Day (Time)	Measurement	RIMPUFF	HySplit
27/09/2008 (16:20)	25.5 ± 5.5	502.3	0
28/08/2008 (02:00)	11.2 ± 2.8	0.9	13.3

29 – 30th September

Day (Time)	Measurement	RIMPUFF	HySplit
29/09/2008 (23:09)	229 ± 16	4.6	0
30/09/2008 (05:49)	7.7 ± 1.5	0.1	1.9
30/09/2008 (06:53)	13.8 ± 3.1	0.5	30.7
30/09/2008 (09:03)	43.2 ± 16.8	12.1	0

1st October

Day (Time)	Measurement	RIMPUFF	HySplit
01/10/2008 (09:19)	4.5 ± 1.9	0	0
01/10/2008 (10:38)	6.6 ± 1.2	0	0

2nd October

Figure 4. Sampling locations, and significant [85]Kr results, for the DISPAMER campaign (27th Sep– 2nd Oct 2008)
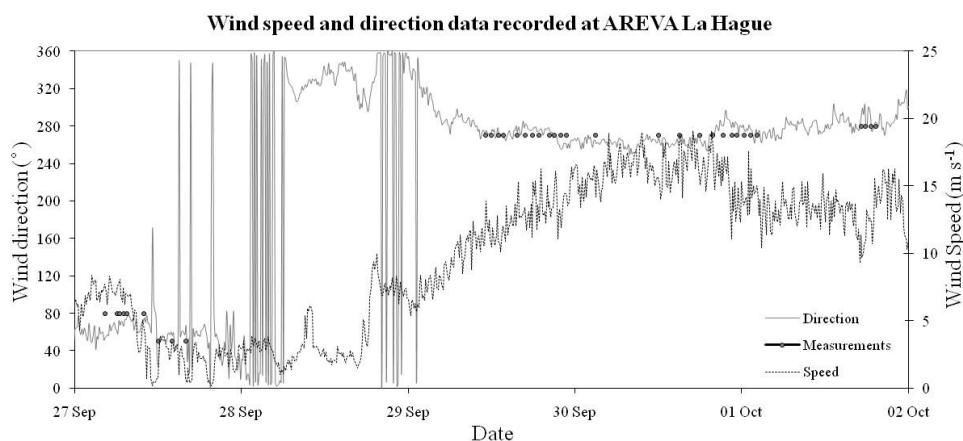

Figure 5. Wind speed and direction data recorded at AREVA La Hague during the DISPAMER campaign.

Of the 40 samples analysed, only eight measurements (Figure 4) were above 4 Bq m^{-3} (almost twice background). All others results varied between 1.4 ± 0.4 and 3.4 ± 2.6 Bq m^{-3}. As mentioned above, the DISPAMER campaign took place during a period of poor weather conditions (high wind speeds and fast changing wind direction/speeds, Figure 5). The weather conditions persisting over the measurement campaign are unlikely to have been accurately predicted by the meteorological data used in the models. In general, the situations where

the models incorrectly predict ^{85}Kr values are most likely a result of the low temporal resolution of the meteorological data files.

The model predictions for the DISPAMER campaign were compared to measured concentrations for a single point (in location and time). In this regard, it is important to note that it is difficult to achieve perfect agreement between models and measured concentrations even during ideal weather conditions. RIMPUFF and HySplit apply (numerical) approximations in order to simulate atmospheric dispersion processes. This leads to uncertainties in the results. Further uncertainties may arise from the meteorological data, the meteorological pre-processor, the terrain data, etc. These uncertainties are compounded and amplified when comparing values for a single location/time.

5. CONCLUSIONS

The ^{85}Kr measurement methods developed by IRSN have been shown to be reliable up to distances of several tens of km from the source to measure ^{85}Kr activity concentrations in air to levels as low as atmospheric background (1.5 Bq m^{-3}). The measurements presented in this paper indicate that ^{85}Kr is a good tracer of atmospheric dispersion processes and is potentially useful in the validation of ATDMs.

In this work, the RIMPUFF and HySplit models made reasonable estimates of the ^{85}Kr measurements for the land based sampling sites (Doville and LRC) and were able to reproduce the overall temporal shape of the ^{85}Kr concentrations. The models failed to correctly reproduce concentration peaks during periods of rapid and large variations in wind speed and/or direction. This underperformance is related to the time resolution of the meteorological data that drive the models: the time steps are too large to account for the rapid variations in wind speed and direction when they occur. The ADMS model, in its simplest configuration cannot reproduce the experimental measurements at mesoscale distances.

Acknowledgements

References

[1] Kok Y.S., Eleveld H., Schnadt H., Gering F., Gregor, J., Böttge H. and Salfeld C., *Radiat Prot Dosim* **113** (2005) 381-391.
[2] Harrison R.M., ApSimon H.M., Beck H., Dickerson D., Garland J.A., Graziani G., Gudiksen P., McKay W.A., Mishra U.C., Nicholson K.W., Smith F.B. and Shapiro, C.S., 1993. "Atmospheric Pathways", SCOPE 50 Radioecology after Chernobyl, F. Warner & R.M. Harrison Eds. (John Wiley & Sons, Chichester, 2003) 55-100.
[3] Cooper J.R., Randle K. and Sokhi R.S., 2003. Radioactive Releases in the Environment: Impact and Assessment (John Wiley & Sons, Chichester, 2003).
[4] Ahlswede J., Hebel S. and Kalinowski M.B., Ross O., Update of the global Krypton-85 emission inventory: Occasional paper N°9 (University of Hamburg, Hamburg, 2009).

[5] Gurriaran R., Maro D., Bouisset P., Hebert D., Leclerc G., Mekhlouche M., Rozet M. and Solier L., *J Environ Radioactiv* **72** (2004) 137-144.
[6] Draxler R.R. and Hess G.D., Description of the HySplit 4 modelling system. (NOAA Technical Memorandum ERL ARL-224, Maryland, 2004)
[7] CERC, 2009. *ADMS 4.0 User Guide*, Cambridge Environmental Research Consultants, (2009).
[8] Thykier-Nielsen S., Mikkelsen T. and Moreno J., *Radiat Prot Dosim* **50** (1993) 249-255.